# A CHEMICAL STUDY OF OILS AND FATS

# OF ANIMAL ORIGIN

# A CHEMICAL STUDY OF OILS AND FATS OF ANIMAL ORIGIN

# BY M. E. CHEVREUL

You have to strive to be infallible without claiming to succeed Malebranche

Translated and annotated by Albert J. Dijkstra

Edited by Gary R. List and Jaime Wisniak

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# NICOLAS-LOUIS VAUQUELIN,

MY TEACHER

## TABLE OF CONTENTS

# TABLE OF CONTENTS

Contributors	xix
PREFACE (G.R. List)	xxi
BIOGRAPHY OF M.E. CHEVREUL (J. Wisniak)	xxiii
FOREWORD BY THE TRANSLATOR/EDITOR (A.J. Dijkstra)	xxvii
INTRODUCTION	xxxi

## BOOK I

CHAPTER 1 Definitions	1
Definition of species as present in a mixture	1
Definitions of the words variety, genus, immediate principle	2
The term <i>oils and fats</i> cannot be the subject of a scientific definition	3
CHAPTER 2 Description of the analytical method to determine the	
elemental composition of oils and fats	5

# BOOK II

TABLE LISTING SPECIES OF LIPIDS DESCRIBED IN THIS WORK	13
CHAPTER 1 Stearic acid and some stearates	14
SECTION 1 Stearic acid	14
§1 Composition	14
§ 2 Physical properties of free stearic acid	15
§ 3 Chemical properties that are observed without the acid being	
altered	16
§ 4 Chemical properties that are observed when the acid is altered	16
§ 5 Occurrence	20
§ 6 Preparation	20
§7 Nomenclature	20
§8 History	20
SECTION 2 Stearates	21
ART. 1 Potassium stearate	21

ART. 2 Potassium bistearate	24
ART. 3 Sodium stearate	30
ART. 4 Sodium bistearate	31
ART. 5 Barium stearate	32
ART. 6 Strontium stearate	32
ART. 7 Calcium stearate	33
ART. 8 Lead stearate	33
ART. 9 Basic lead stearate	34
ART. 10 Ammonium stearate	34
CHAPTER 2 Palmitic acid and some palmitates	41
SECTION 1 Palmitic acid	41
§1 Composition	41
§ 2 Physical properties	42
§ 3 Chemical properties that are observed without the acid being	
altered	42
§ 4 Chemical properties that are observed when the acid is altered	42
SECTION 2 Palmitates	44
ART. 1 Potassium palmitate	44
ART. 2 Potassium bipalmitate	44
ART. 3 Sodium palmitate	45
ART. 4 Sodium bipalmitate	46
ART. 5 Barium palmitate	47
ART. 6 Strontium palmitate	47
ART. 7 Calcium palmitate	48
ART. 8 Lead palmitate	48
ART. 9 Basic lead palmitate	48
ART. 10 Ammonium palmitate	49
Observations concerning stearic and palmitic acid	49
Observations concerning stearle and paintitle acte	<b>H</b> )
CHAPTER 3 Oleic acid and some oleates	54
SECTION 1 Oleic acid	54
§1 Composition	54
§ 2 Physical properties	55
§ 3 Chemical properties that are observed without the acid being	
altered	55
4 Chemical properties that are observed when the acid is altered	56
§ 5 Occurrence	57
§6 Preparation	57
§7 Nomenclature	57
§8 History	57
Melting points of mixtures of oleic acid and palmitic acid (table)	58

SECTION 2 Oleates	
ART. 1 Potassium oleate	
ART. 2 Potassium peroleate	
ART. 3 Sodium oleate	
ART. 4 Barium oleate	•
ART. 5 Strontium oleate	•
ART. 6 Calcium oleate	•
ART. 7 Magnesium oleate	
ART. 8 Zinc oleate	
ART. 9 Copper oleate	
ART. 10 Cobalt oleate	
ART. 11 Nickel oleate	•
ART. 12 Chromium oleate	•
ART. 13 Basic lead oleate	•
ART. 14 Ammonium oleate	
1 <sup>st</sup> Table: Comparative examination of sodium stearate from mutton	
tallow, sodium palmitate from human fat and sodium oleate	
from human fat	•
CHAPTER 4 Phocenic acid and some phocenates	•
SECTION 1 Isovaleric acid	
§1 Composition	
§ 2 Physical properties.	•
§ 3 Chemical properties that are observed without the acid being	
altered	
§ 4 Chemical properties that are observed when the acid is altered	
§ 5 Preparation § 6 Nomenclature	
§ 7 Occurrence and history	
§7 Occurrence and history	•
SECTION 2 Isovalerates	
ART. 1 Barium isovalerate	
ART. 2 Strontium isovalerate	
ART. 3 Calcium isovalerate	
ART. 4 Potassium isovalerate	
ART. 5 Sodium isovalerate	
ART. 6 Lead isovalerate	
ART. 7 Basic lead isovalerate	

CHAPTER 5 Butyric acid and some butyrates
SECTION 1 Butyric acid
§1 Composition
§ 2 Physical properties
\$3 Chemical properties that are observed without the acid being
altered
4 Chemical properties that are observed when the acid is altered
§ 5 Nomenclature
§ 6 Occurrence and history
<u> </u>
SECTION 2 Butyrates
ART. 1 Barium butyrate
ART. 2 Strontium butyrate
ART. 3 Calcium butyrate
ART. 4 Potassium butyrate
ART. 5 Sodium butyrate
ART. 6 Lead butyrate
ART. 7 Basic lead butyrate
ART. 8 Copper butyrate
ART. 9 Zinc butyrate
ART. 10 ammonium butyrate
SECTION 1 Caproic acid
§ 2 Physical properties.
§ 3 Chemical properties that are observed without the acid being
altered
§ 4 Chemical properties that are observed when the acid is altered
§ 5 Preparation
§ 6 Nomenclature
§7 Occurrence.
§8 History
SECTION 2 Caproates
SECTION 2 Caproates
1
L
ART. 4 Potassium caproate
ART. 5 Sodium caproate
ART. 6 Ammonium caproate
CHAPTER 7 Capric acid and some caprates

SECTIC	N 1 Capric acid	113
	Composition	113
§ 2	Physical properties	114
<b>§</b> 3	Chemical properties	114
	Preparation	114
	Nomenclature	115
	Occurrence	115
	History	115
U		-
Sectio	ON 2 Caprates	116
	ART. 1 Barium caprate	116
	ART. 2 Strontium caprate	117
	-	
CHAPTER	8 8 Hircic acid	119
CHAPTER	89 Cholesterol	120
	Composition	120
	Physical properties	120
	Chemical properties that are observed without the cholesterol	
U	being altered	120
<b>§</b> 4	Chemical properties that are observed when the cholesterol is	
0 -	altered	121
8.5	Preparation	123
	Nomenclature	123
	History	123
U	5	
CHAPTER	R 10 Ethal	126
§1	Composition	126
	Physical properties	126
<b>§</b> 3	Chemical properties that are observed without the cetyl	127
	alcohol being altered	
$\S4$	Chemical properties that are observed when the cetyl alcohol	129
_	is altered	
§ 5	Preparation	130
	Nomenclature	130
	History	131
CHAPTER	R 11 Cetin	132
§1	Composition	132
	Physical properties	132
	Chemical properties that are observed without the	
-	spermaceti being altered	132
§ 4	Chemical properties that are observed when the spermaceti	
-	is altered	132
§ 5	Preparation	135
	History and nomenclature	135
0 -	5	-

CHAPTER	12 Mutton tallow stearin
§1	Composition
	Physical properties
§ 3	Chemical properties that are observed without the stearin
	being altered
<b>§</b> 4	Chemical properties that are observed when the stearin is
0 -	altered
\$5	Occurrence
e	Preparation
	Nomenclature
	History
30	
Снартер	13 Human fat stearin
	Composition
	Physical properties
	Chemical properties that are observed without the stearin
30	being altered
S 1	Chemical properties that are observed when the stearin is
51	altered
85	Occurrence
30	
CHAPTER	14 Olein
	Composition
	Physical properties
	Chemical properties that are observed without the olein
80	being altered
S /	0
84	Chemical properties that are observed when the olein is
с <b>г</b>	altered
-	Occurrence
	Preparation
	Nomenclature
§ 8	History
Снарты	215 Phocenin
	2 15 Phocenin
	Physical properties
	Chemical properties
	Preparation
	Nomenclature
§ 5	History
CHADTER	216 Buturin
	Physical properties
	Physical properties
	Chemical properties
	Preparation
	Nomenclature
§ 5	History

CHAPTER 17 Hircin	149
-------------------	-----

# **BOOK III**

PREPARATION OF SPECIES OF FATTY MATERIALS	151
CHAPTER 1 Preparation of fatty acids and analysis of the saponification products of the fatty material of the 5 <sup>th</sup> and 6 <sup>th</sup> genera	151
<ul> <li>SECTION 1 Preparation of the fatty material to be saponified</li> <li>§ 1 Preparation of human fat, lard, beef tallow, mutton tallow, etc.</li> <li>§ 2 Preparation of dolphin or porpoise oil</li> <li>§ 3 Preparation of butter</li> </ul>	152 152 152 152
SECTION 2 Saponification by caustic potash of fats, dolphin or porpoise oil and butterfat	153
SECTION 3 Analysis of the above saponification products §1 Analysis of the saponified fatty material and preparation of	154
the constituent fatty acids ART. 1 Analysis of the fatty material of soap of human fat,	154
consisting of palmitic and oleic acids ART. 2 Analysis of the fatty material consisting of stearic, palmitic and oleic acid originating from soaps made	154
from mutton tallow, beef tallow and lard § 2 Examination of the aqueous liquid; preparation of glycerin	157
and volatile acids	159
ART. 1 Preparation of isovaleric acid	160
ART. 2 Preparation of the acids present in butter	162
Preparation of butyric acid	172
Preparation of caproic acid	174
Preparation of capric acid	174
Preparation of hircic acid	175
CHAPTER 2 Preparation and saponification of spermaceti	182
§1 Preparation of spermaceti §2 Analysis of the saponification products of spermaceti and the	182
preparation of cetyl alcohol	183
Formula for separating the various products arising during the treat-	4.04
ment of fatty material that can be saponified by potassium hydroxide	191

# **BOOK IV**

COMPAR	RATIVE STUDY OF VARIOUS KINDS OF FATS AND FATS FROM	
CADAVE	RS	19
Снарте	R 1	19
	Various properties of fats that can be distinguished in fats	19
50	without decomposing them	1
82	Changes in nature when the fats react with potassium hydroxide	1
83	Comparative study of the solid fatty acids of various kinds of	1
30	fats	1
	ART. 1 The pearly material and its acids	1
	ART. 2 Oleic acid	1
84	Comparative study of the stearin and the olein of various fats	1
31	ART. 1 Stearins	1
	ART. 2 Oleins	2
\$5	Conclusions drawn from the facts reported in the four previous	-
50	paragraphs	2
	r	
CHAPTE	R 2 <i>Study of cow's milk and preparation of butyrin</i>	2
Secti	ON 1 Separation of the fatty part of butter from buttermilk	2
Secti	ON 2 Butter properties and their determination	2
	Properties that can be observed in butterfat without separation	
-	of its immediate principles	2
§ 2	Saponification of butterfat with caustic potash	2
	Analysis of butterfat with alcohol	2
	ART. 1 Aqueous liquids Nos. 1 and 2 and the alcohol used	
	to analyze the butterfat	2
	ART. 2 Butterfat stearin	2
	ART. 3 Oil No. 1	2
	ART. 4 Oil No. 2	2
Secti	ON 3 The preparation of butyrin	2
CHAPT	ER 3 Study of several cetacean oils	2
Secti	ON 1 Examination of the oil of the Harbor porpoise	2
	Preparation	2
	Properties of the oil	2
	Analysis of the oil with alcohol and preparation of phocenin	2
-		
Secti	ON 2 Study of the oil of <i>Delphinus globiceps</i>	2
§1	Preparation	2

§ 2 Properties of the oil	222
§ 3 Chemical analysis of the <i>Delphinus globiceps</i> oil by exposing it to the cold	222
ART. 1 Examination of the crystallized substance	223
ART. 2 Examination of dolphin oil from which the spermaceti crystals had been removed	224
SECTION 3 Examination of fish oil	226
ART. 1 Examination of the liquid part of fish oil	226
ART. 2 Examination of the solid part of fish oil	227
CHAPTER 4 Examination of cadaver fat and adipocere	231
§1 Analysis of the fat extracted from a cadaver from the Cemetery	
of the Innocents	231
ART. 1 Examination of the alcohol-insoluble residue ART. 2 Examination of the deposit formed during washing	232
with alcohol	233
ART. 3 Examination of the alcoholic washing liquors from	
which the above deposit was separated	233
ART. 4 Conclusions from the previous examinations	234
§ 2 Examination of adipocere	234
ART. 1 Examination of the pearly material from adipocere	
soap	236
ART. 2 Examination of the oily acid	236
ART. 3 Examination of the aqueous mother liquor of the	
adipocere soap	236
ART. 4 Conclusions.	236
§ 3	238
ART. 1 Bone marrow partially converted into fat	238
ART. 2 Ram fat formed in the middle of a pool of stagnant	
water containing calcium sulfate and carbonate	239

## BOOK V

INTRODUCTION	243

# PART ONE

Saponification considered with respect to the fatty materials	244
CHAPTER 1	245
§ 1 Is any acetic acid produced during saponification?	245
§ 2 Is any carbon dioxide produced during saponification?	245
§ 3 Is oxygen necessary for saponification?	247

CHAPTER 2 Some general observations regarding saponification		
CHAPTER 3 On saponification	252	
SECTION 1 The saponification of mutton tallow, lard and human fat § 1 Comparative elemental analysis of mutton tallow, lard and	253	
human fat	253	
§ 2 Comparative elemental analysis of the saponification products		
of mutton tallow, lard and human fat	254	
Elemental analysis of the free fatty acids	255	
Comparative elemental composition of the anhydrous fatty acids	255	
Analysis of glycerin	256	
Table 1 Saponification of mutton tallow	257	
Table 2 Saponification of lard	259	
Table 3 Saponification of human fat	258	
Results and conclusions	260	
SECTION 2 The saponification of spermaceti	263	
Table The saponification of spermaceti	265	
SECTION 3 Reflections on the reasons why cholesterol is not		
saponified	267	

### PART TWO

Saponification with respect to bases that can form salts		
CHAPTER 4 The effect on lard of several bases that can form salts	271	
§ 1 The effect of caustic soda on lard	271	
§ 2 Saponification of the fat by baryta	272	
§ 3 Saponification by strontium oxide	273	
§ 4 Saponification by lime	273	
§ 5 The effect of magnesia on lard	273	
§ 6 The effect of ammonia on lard	274	
§ 7 The effect of alumina on lard	275	
§ 8 Saponification by zinc oxide	276	
§ 9 The effect of copper oxide on lard	276	
§ 10 Saponification by lead oxide	277	
Summary of Chapter 4	278	
CHAPTER 5 On the amount of fat that a given amount of potassium	281	
hydroxide can saponify	201	
CHAPTER 6 Can saponification be realized by potassium bicarbonate and ammonium carbonate?	284	
§ 1 The effect of potassium bicarbonate on lard	284	

285
286
287
289

# **BOOK VI**

SUMMARY AN	ID GENERAL REFLECTIONS	293
SECTION 1	Knowledge of oils and fats up to 1813	294
SECTION 2	General views on the immediate composition of oils and fats	295
SECTION 3	Preparation of fatty species	300
SECTION 4	Various properties regarding solvents of potassium and sodium stearates, palmitates and oleates	308
SECTION 5	General views on the interaction between bases that can form salts and oils and fats	313
SECTION 6	Applications of my research	317
Section 7	Conjectures on the composition of various species of fatty materials	322

## NOTES

FIRST NOTE	On the analysis of goat's milk butter	331
SECOND NOTE	The effect of oxygen on lard	332
	The effect of sulfuric acid on stearins and olein e effect of sulfuric acid on lard e effect of sulfuric acid on mutton tallow stearin	334 334 337
Section 1 Th	e effect of sulfuric acid on phocenin e effect of sulfuric acid on butyrin	339 339 340
ILLUSTRATIONS		343
INDEX		345

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#### Contributors

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In 1978, he became involved in edible oils and fats when he joined the Vandemoortele Group in Izegem, Belgium as R & D Director. He is the inventor in a dozen patents and has published numerous articles and book chapters on edible oil processing; he has also acted as co-editor.

He was the first non-American to receive the American Oil Chemists' Society (AOCS) Chang Award (1997) and also the first to receive the European Lipid Technology Award (2002). In 2009, he was selected for the Alton E. Bailey medal of the AOCS and the Chevreul medal of the Association française de l'étude des corps gras (AFECG). Although officially retired, he continues to be active in the field of edible oils and fats as an author, editor, inventor and scientific consultant.

**Gary R. List** was a research chemist at the Northern Regional Laboratory (USDA) in Peoria, Illinois, from 1963-2007. His research involved edible oils, oilseed processing and fat modification. He is the author of over 310 publications and has edited 4 books on fats, oils and lipids. His honors/awards include the Bailey Medal, Fellow and Award of Merit of the American Oil Chemists' Society (AOCS), and the Chang, Macy, and Division Lecture Awards of the Institute of Food Technologists (IFT). He is also the recipient of the Outstanding Achievement Award of the United Soybean Board and the European Lipid Technology Award (2009). While retired, he continues to be active in society work and consulting for the oilseed industry.

Jaime Wisniak, Ph.D. was born in Santiago, Chile. He has a degree in Chemical Engineering (1957), Pontificia Universidad Católica de Chile (PUC); PhD in Chemical Engineering, Purdue University, (1960); B.Sc. in Chemistry Ben-Gurion University of the Negev (BGU), Israel (1987); and B.Sc. in Life Sciences, BGU, Israel (1992). Between 1960 and 1970 he was a member of the Faculty of Engineering at PUC, where he served as Head of the Department of Chemical Engineering, Director of the School of Engineering, and Director of the Plan of Development. In 1971 he joined the Department of Chemical Engineering of BGU.

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#### Preface

In 1811, when Michel Chevreul began his research, the chemical nature of fats, oils and lipids was unknown. Some 12 years later he published his classic work based on a series of publications that appeared in *Annales de chimie* during the period from 1813-1823. This work entitled: "*Recherches chimiques sur les corps gras d'origine animale*" is of interest not only for establishing the chemical nature of fats but also from a historical point of view by giving an insight into the mind of one of the premier organic chemists of the early 19<sup>th</sup> century. Berzelius commented that Chevreul's work should serve as a model for young chemists wishing to do research in any field of chemistry and went on to say that Chevreul's investigations were the most complete and best executed series of experiments in all of chemistry.

Von Liebig also had glowing comments about Chevreul's work: "Chevreul has shown the connection between the saponification products and their mode of formation and that work was the starting point of all subsequent research and the basis for all prevailing methods of analysis and investigations of organic materials." We must thank Chevreul for the beautiful principles underlying every organic investigation. Other contemporaries declared: "Chevreul's work is a masterpiece."

It is therefore most surprising that this masterpiece has never been translated into English. It was reprinted in 1825, in 1886 on the occasion of the author's 100<sup>th</sup> birthday and again in 1986. It has also been published as a pdf-file and can be accessed via <u>http://books.google.com</u> but all these publications are in French, which limits their accessibility; a translation into English is therefore long overdue. Accordingly, the American Oil Chemists' Society has taken the laudable initiative of distributing our translation of this masterpiece on the occasion of its own centenary.

However, translating this book is complicated not only because it is written in 19<sup>th</sup> century French but also since it requires knowledge of contemporary 19<sup>th</sup> century chemistry. Dr Albert Dijkstra has responded to both of these challenges by translating the text into a more modern and readable form and by annotating the work in the light of modern chemical knowledge. The translator and the editors hope the reader will enjoy this remarkable contribution to the history of lipid chemistry.

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Héliog. Dujardin.

M.E.CHEVREUL 31 Août 1886.

E cherrent

Imp. Eudes.

### **Biography of M.E. Chevreul**<sup>\*</sup>

Michel Eugène Chevreul (1786-1889) is an excellent example that old age is not necessarily an impediment to a fruitful productive life. He lived to be almost 103 years and to the very last minute was very active in French science. His contributions to the chemistry of fats and to the chemistry and physics of color were fundamental to the understanding of the principles that govern them. His discovery of the chromatic circle put fabric dyeing on a scientific basis that led to the flourishing development of tapestry, and his discoveries related to fatty materials such as stearin put the candle industry on the road to its modern development. Chevreul established the nature of fats and their saponification, and isolated and described many of the commonly known fatty acids.

His name is not one that appears in standard books of chemistry and physics. No reaction or theory carries his name but his fundamental and applied research stands behind many common products like drying oils, soaps and candles. His work signals the understanding of the chemistry of fats.

When Chevreul was old enough to go to school, all educational institutions had been disbanded, and for five years his parents had to have recourse to private teachers. In 1803 he moved to Paris to pursue his chemical interests in the laboratory of Louis Nicolas Vauquelin (1763-1829) at the Muséum d'Histoire Naturelle. His first activities under Vauquelin were the study of the coloring materials present in natural products. Between 1807 to 1811 he was able to isolate brazilin from brazilwood, hematin from logwood, quercitrin from oak bark, luteolin from dyer's wood, and morin from fustic.

Chevreul was elected to the Académie des Sciences in 1826 and in 1830 he was appointed to the chair in chemistry at the Muséum, in replacement of Vauquelin. Chevreul continued to work and attend the Académie meetings until he was 102. On the jacket of Bouchard's book it is written that Chevreul witnessed in his own country four kings, two emperors, three republics, and four revolutions. When he was about eighty-five years old, instead of adding some of his professional and academic titles to his name on the title pages of his publications, he wrote simply "*M. Chevreul, doyen des étudiants de France.*" He read his last communication to the Académie in May 1888. He died April 9, 1889, five months before the Eiffel tower was inaugurated. His state funeral at the Cathedral of Notre Dame attracted thousands of people who massed near the church in spite heavy rains. Chevreul's ashes are buried at the cemetery of l'Hay.

<sup>&</sup>lt;sup>\*</sup> Taken from Wisniak, J., Michel Eugène Chevreul, Educ. Quím., **13**(2), 133-141 (2002).

Chevreul was a pioneer not only in organic chemistry but also in many other fields. The results of his experimental work were not sufficient and his curiosity led to his contributions in the areas of dyeing, color theory, and psychology. Many of the questions that he asked himself could not be answered experimentally, but only by patient efforts in the library and long meditations. His appointment, in 1824, as director of dyeing at the Manufacture Royale des Gobelins, the national tapestry works of France, marked a new direction in his career. Chevreul realized immediately that dyeing was done according to recipes and that to improve the process it was necessary to put it on a scientific basis. He set up an experimental program to determine the character and properties of fibers, the mutual actions occurring between fibers and acids, bases, and salts, between fiber and organic dyes, and between combinations of these. He also analyzed the influence of mordants on the tone and stability of colors, bleaching, and preservation of colors, and the effect of water quality on the finished material. In order to solve the problem of the definition and representation of colors Chevreul proceeded to define a chromatic circle in which, out of three pigments of red, yellow, and blue, he was able to realize almost fifteen thousand shades or tones.

Chevreul's investigations of the chemistry and composition of fats and oils constitute his most remarkable scientific contribution. He was led into this subject in 1811 when Vauquelin asked him to analyze a sample of a potassium soap made from pig fat. As an initial step Chevreul prepared a very dilute aqueous solution of the soap and observed that shiny crystals separated from it (matière nacrée). Acid treatment of this fraction of the soap, followed by crystallization in alcohol, yielded a solid fatty substance that possessed the properties of an acid, a property that was in contrast to the neutral fat from which the soap was derived. He named this fatty substance margarin. In a later work he changed the name margarin to margaric acid. Margaric acid (although misidentified as a pure acid) was the first of the several fatty acids that he was to isolate from natural fats. In a following paper Chevreul reported that from the dissolved part of the soap he had obtained a second acidic product, this one of a fluid character. He named it *fluid fat*, later redesignating it *oleic* acid. Having determined the results of the saponification process, Chevreul went on to determine whether the products of saponification were originally present in the fat. His results indicated that the fat was neutral before saponification and that after combination with alkali it produced the solid and liquid fatty acids and the sweet principle of Scheele. In a later work Chevreul named the sweet principle glycerin. In a few words, Chevreul elucidated the process of the formation of soap and showed that fats are combinations of acids with glycerin.

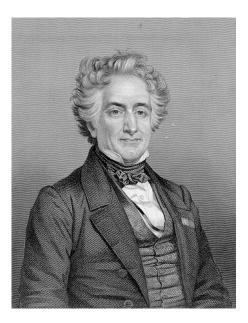
Chevreul studied the saponification process using a variety of bases; he saponified fats from all available sources, including man, wild and domesticated animals, oils of vegetable origin, and the crystalline substance present in biliary calculi (cholesterol). Such origins are revealed in the names he coined for the new acids: *butyric* (butter); *capric*, *caproic* (goat); *delphinic* (dolphin); *phocenic* (porpoise); etc. His investigation of the nature of spermaceti led him to purify cetin by repeated crystallization. Saponification of spermaceti led him to prepare cetyl alcohol.

Chevreul utilized his discoveries to develop a system of nomenclature for fats and derived products. The chemical substance in biliary calculi he called *cholesterin*; spermaceti was to be *cetin*, the solid fat principle and the liquid fat principle were to be *stearin* and *elain* (oil; eventually he would change this name to *olein*). The acid from saponified spermaceti was named cetic acid and the terms margarates, oleates, and cetates, were to be the generic names for the soaps, which these acids formed with bases. His copious research on the subject eventually led to a 500-page book summarizing the state of the art in the chemistry of fats.

Chevreul's experiments on the saponification of fats led him to believe that the product could be used for manufacturing new and improved candles: together with Joseph-Louis Gay-Lussac (1778-1856) they financed the building of a factory for the manufacture of stearic acid by treating fats with alkali or acid. Although this venture was a commercial failure, the further development of the French industry for the manufacture of candles from fatty acids is based on Chevreul's work. In recognition of these merits, Chevreul was awarded in 1852 the Marquis d'Argenteuil prize for the encouragement of national industry (12 000 francs). The jury of the International Exposition of Paris of 1855 also recognized these merits and awarded him a grand medal of honor.

Besides chemistry and physics, Chevreul was also interested in psychology and the history of science. In 1853 he was head of a committee appointed by the Académie to inquire into the phenomena of the divining rod and the pendulum, which had been the subjects of recent memoirs to the Académie and were thought to be the result of the organoelectricity, which resided in humans. Chevreul experimented with the pendulum and explained it as the result of expectant suggestion; the swinging pendulum was a case of autosuggestion: as long as you believed the movement possible, it occurred. Chevreul's incursion into psychology led him to write a book on scientific method where he stated that he always sought to establish general methods and principles and was never satisfied with making individual discoveries. He claimed that one of the principal functions of experiments was the testing of the validity of preliminary conclusions based on the study of natural phenomena. All generalizations should be submitted to rigorous verification by experiments and observation.

In 1866 Chevreul published the first volume of his history of chemical knowledge, which was meant to include four volumes. In the first volume he discussed the purpose and field of chemistry, its relationship with other sciences, with natural history and with life itself; the classification of the sciences; and scientific abstraction versus art and literature.



M.E. Chevreul at 50 years of age; lithograph by Maurin; engraving by Conrad Cook

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#### Foreword by the translator/annotator

When working on a text, a translator should try to keep as close to the original as possible and try to make sure that people reading the translation get an identical impression as those able to read the original. If the author expresses himself ambiguously, this ambiguity should be maintained. Even mistakes should be preserved, although the translator can insert a footnote pointing out this mistake and thereby avoid being blamed for it.

This means that there are two kinds of footnotes: those by the author and those by the translator. The author numbered his footnotes per page and started again with (1) on every new page. Accordingly a distinction between these two kinds of footnotes will be introduced by keeping the author's footnotes on the page where they arise and identifying them by symbols like: \*, †, ‡ and §. Footnotes by the translator/ annotator will be numbered consecutively and listed in numerical order at the end of each chapter as endnotes.

Like authors, languages also differ in style. Some aim to be very precise whereas others are less finicky. Some glory in being ornate while others prefer to avoid flowery expressions. Accordingly, translating an ornate sentence literally into a language that prefers simplicity amounts to rubbing the reader up the wrong way. He won't like it since what he is forced to read is evidently translated. So whereas the translator should aim to maintain the style of the author, he (or she) should also adapt his style to the language he is translating into.

Accordingly, the title of the present work could be translated into modern English as: *The Chemistry of Animal Fats*. That is much shorter and more direct than a more literal translation such as *A Chemical Study of Oils and Fats of Animal Origin*. Moreover, the left page header uses a running title *LES CORPS GRAS D'ORIGINE ANIMALE*, presumably because there was no room for the full title. So a choice had to be made: For the full title of the book, the long translation was chosen since it may remind people of the original. The running title chosen is ANIMAL OILS AND FATS.

Editors face a different task. When they edit a book consisting of contributions from different authors, their prime task is to avoid overlap and ensure some degree of internal consistency. However, when editing a translation, their role is more limited. They can supply background information and comment on and/or suggest further endnotes and assist the copy editor in his efforts to maintain internal consistency.

What about a translator who works on a book that was published in 1823 and is of the greatest historical significance? In that case, I think it is the moral duty of the translator/annotator to make sure that above all, the historical significance of the book is highlighted. Consequently, he should feel free to modernize the style of writing by for instance chopping up sentences that in the original cover more than half a page, and cut out some verbosity. He should introduce modern technical jargon and explain this in an endnote. And above all, he should concentrate on making sure that the scientific achievements of the author are well explained and put in their historical context.

Palmitic acid (C16:0 or hexadecanoic acid) was discovered by Chevreul. Because it formed nice pearly crystals, he called it "acide margarique" after the Greek word for pearl: μαργαρίτης. Nowadays, margaric acid refers to a different fatty acid: heptadecanoic acid (C17:0). Accordingly, the name 'palmitic acid' will be used for the acid the author refers to as 'acide margarique'.

Similarly, ancient names of various chemical reagents will be replaced by their current, more systematic names. In his experiments, Chevreul regularly uses "massicot" and I wonder just how many Frenchmen today know that this compound is actually lead oxide, which they would now refer to as "oxyde de plomb". So, although there is an equivalent ancient word in English, I use 'lead oxide', since this is the term in current use.

In another respect, the author was very modern in that he used the metric system of measurement as adopted by the French government on 7 April 1795. He uses degrees centigrade which he just calls 'degrees'; they have been translated as °C. He expresses densities as grams per milliliter but does not mention this unit of measurement. Where the author mentions a unit, this has been mentioned in the translation. If he has not mentioned the unit concerned, it has been added in brackets in the translation. If he describes properties in a different way than is customary today, an endnote provides the conversion.

When trying to understand what the author meant, I have made extensive use of the worldwide web. In fact, I would not have been able to understand many of his expressions without access to the web. At a certain stage, Chevreul mentions "potasse à l'alcool" which at first sight looks like an alcoholic solution of potassium hydroxide. However, when consulting the web, I learned that potassium hydroxide was produced by the causticization of a solution of potassium carbonate according to:

$$K_2CO_3 + Ca(OH_2) \rightarrow 2 \text{ KOH} + CaCO_3 \downarrow$$

If then the supernatant solution of potassium hydroxide is evaporated to dryness, it tends to absorb carbon dioxide from the air and thus yield an impure product. The way to get a less impure product involved mixing the concentrated potassium hydroxide solution with alcohol. This led to the formation of two phases: an upper alcoholic phase with reasonably pure potassium hydroxide and a lower, aqueous phase with potassium carbonate and other salts. In other words, what I originally intended to translate as 'alcoholic solution of potassium hydroxide' turned out to contain no alcohol. This is just an example of getting to grips with the subject and of arriving at a solution. For the annotator, doing this has been a constant source of enjoyment, just as appreciating the many contributions the author made to lipid chemistry.

In one way, the layout used by Chevreul or his publisher was extremely modern: he numbered his sub-sections just as US Patent specifications started to do only a few years ago. However, Chevreul had a reason to do so since he used these numbers to refer to these subsections; I have therefore kept these numbers. Another way in which the author facilitated browsing through his book was by having side notes printed in the margin. I found them useful so I have retained them as well.

Because this translation has been published by printing-ondemand, I was also responsible for the copyediting. To this end and at the instigation of AOCS Press, I have made extensive use of *The Chicago Manual of Style* but at the same time, the style of the original work has been maintained whenever possible.

Finally, I want to express my thanks to Gary List for taking the initiative of commissioning the English translation of the first book ever published on lipochemistry; to Jaime Wisniak for suggesting background literature, the inclusion of an index and ways to improve my first efforts at translating this monograph; and to the AOCS Press for fulfilling a public service by distributing it. Above all, I want to express special thanks to my wife Liz for undertaking the mammoth task of correcting my translation and polishing my English, which for me still is a foreign language, just like French. Her contributions have been very necessary, highly professional and extremely valuable.

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#### INTRODUCTION

When undertaking a chemical research project that is likely to expand, a scientist will almost always start with a random sequence of observations. So when reporting the results in the order in which he found them, he will be following a course that he has not consciously chosen. If on the other hand, he could have postponed the presentation of his findings, he would have been able to oversee them as a whole and present his findings in the order he felt to be the most suitable. But now he has to keep repeating himself, either to correct what he said before, to complement his first observations or to emphasize certain aspects that he had originally given little attention. Then there are also the incidental experiments that are nevertheless necessary to elucidate the main research or to substantiate the conclusions drawn from it. There is also the inconvenience of having to give certain substances a circumlocutory designation because we do not yet know enough about them to be able to give them a name. From all this you will realize that most readers will find reading a number of articles on the same subject very tiresome<sup>1</sup> and will therefore tend to take an unfavorable view of work resulting from long observations and numerous experiments.

There are people who are very severe in their judgment of someone who has carried out many experiments on the same subject<sup>2</sup>. They do not forgive him the mistakes he has made and instead of thanking him for clearing up the obstacles and paving the way, they accept what he has done with indifference as if it were the natural result of the time he devoted to his work. They reserve their praise for the man who in a short period of time satisfies their curiosity by presenting a variety of different studies containing just a few interesting facts that can be easily grasped in isolation, since they are not put together to create a coherent system, which is something which always calls for more effort and study to understand and appreciate at its true value. The people we are talking about are not aware that discovering striking but isolated facts is in general much easier than discovering the secondary facts that link them together, since the former are often stumbled on by pure chance. It is when you try to link up these chance observations and arrive at a distinction between the primary facts and secondary consequences<sup>3</sup> that obstacles start mounting up and time seems to pass at a frightening speed for the researcher who cannot see an end to his endeavors. Although this is hard and painstaking work, it nevertheless has indisputable advantages for the advancement of science and for the researcher himself, who finds

Inconvenience of presenting several investigations of the same subject in a series of reports many opportunities to verify the conclusions he has drawn from earlier experiments.

Scientific advantages of a detailed investigation of a subject

Advantages for the scientist of investigating a subject in detail

People say that chemistry consists of facts but there is more to it than just facts. When providing general definitions or classifying compounds according to similar or different properties, known facts must necessarily be interpreted to establish the links between them. Drawing up a definition therefore also means establishing links and assigning different degrees of importance to the various properties. This is more than simple fact-finding, although every interpretation and every definition must be based on well-established facts. Priestley has discovered a large number of important compounds. He has also determined most of their properties and he has certainly provided science with more factual data than Lavoisier. Nevertheless, without wishing to detract from the importance of this great English scientist, we can safely say that posterity attributes more importance to Lavoisier. This can only be because of the way he has interpreted the facts. What holds for the facts that form the basis of the anti-phlogiston theory<sup>4</sup> must also hold for facts that are yet to be discovered. Accordingly, we are of the opinion that it is better for the progress of science if we assemble as many analogous facts as possible and attempt to correlate them so as to arrive at general conclusions which transform a collection of facts into true science, rather than gathering facts on a diversity of unrelated subjects.

If you accept that it is essential to estimate the confidence limits of the experimental data, you will agree that the course we have followed is the most appropriate. If you want to determine the elemental composition of a compound you have recently discovered, various difficulties arise regarding the methods of observation used. Nobody will deny that there can be no discoveries without accurate experimentation. But the art of carrying out experiments, being of a mechanical nature, presents a variety of obstacles to overcome. The results depend on the accuracy of the means or the instruments used. Consequently, it is essential to assess the accuracy of the instruments before using them, whether they be instruments we have inherited from our predecessors or new instruments the nature of the work has obliged us to invent ourselves<sup>5</sup>. Therefore, the investigation of similar but little known compounds necessarily entails a determination of the accuracy of the methods of observation. The large number of experiments he carries out to get to know his oils and fats will provide the scientist with many opportunities to verify his initial observations. This proliferation of experiments will also lead him to take into account differences between the results that he would have passed over or not noticed if he had only performed a small number of experiments, and it will also enable him to disregard certain differences that struck him initially, but which were not confirmed by subsequent repeat experiments. Furthermore, by multiplying the number of angles from which analogous compounds can be looked at, the experiments that are the easiest to carry out can be used to check those that are more difficult to carry out in an accurate manner.

When we started to study natural science, we were struck by the vagueness which then dominated organic chemistry. At the same time, we noted the wide range of applications that this branch of natural science could support in the fields of physiology, anatomy, medicine, and numerous skilled activities and crafts that aim to manufacture useful products for mankind from organic materials. These considerations have long motivated us to reflect on everything connected with the chemistry of organic compounds.

We have applied ourselves to finding out all we could about the immediate principles<sup>6</sup> that can be isolated as such from vegetable and animal matter because *this knowledge forms the basis of organic chemistry and all its applications.* It is in this conviction that we undertook our research into natural and synthetic astringent compounds, minerals and dyes before we started to work on edible oils and fats. It was only after we had made every endeavor to isolate the immediate principles that together constitute the materials under investigation, that we tried to characterize first of all each species by its chemical properties and a range of physical properties, and subsequently to determine the relationships between these species and with the compounds that have already been described.

As we worked to achieve this goal, we also focused our attention on our research tool itself, that is to say on organic analytical chemistry. We tried to understand how the most common reagents work and what influence the major experimental conditions exert when these reagents are allowed to react with organic compounds. For instance, when we studied synthetic astringent compounds, we investigated the effect of sulfuric acid and nitric acid on a fairly large number of compounds. When studying dyes and the extract of woad<sup>7</sup> leaves, we tried very hard to understand how the salts and their immediate principles act in general on the dyes and the organic matter that they precipitate. In the description we published of our distillative autoclave, we considered several questions concerning the use of solvents in organic analytical chemistry and presented a simple method enabling these solvents to act at a higher temperature than is possible when they are heated at atmospheric pressure. Finally, our latest research has made us realize that alcohol, ether<sup>8</sup> and nitric acid do not generate oils and fats when acting upon various compounds containing nitrogen<sup>9</sup>.

With respect to the main experimental conditions used in organic analytical chemistry, we have noticed that the effect of heat on vegetable and animal compounds generates different results depending on whether these compounds are heated in the presence or absence of atmospheric oxygen. We have also noted that alkali acting on many of the same compounds, produces totally different phenomena, depending on whether oxygen is present or excluded. Our observations explain Our motives for studying organic compounds

The way in which we have investigated the composition of organic compounds

How we approached organic analytical chemistry

> 1. Concerning the reagents that are used

2. Concerning the main experimental conditions several anomalies presented by gallic acid, hematin, carmine, etc. when brought into contact with air and alkalis at the same time. We have also realized the influence of oxygen from the atmosphere on the nature of the products resulting from putrefaction. Finally, we arrive at the conclusion that the *decomposition products of the organic compounds that we have examined are less complicated than generally thought. This follows from our observation that we found several immediate principles in matter that has been regarded as pure species. It also follows from our consideration of the influence of air on the action of reagents.* 

Our views on organic analytic chemistry will be expounded in a special monograph. However, when reporting our work, it would be ungrateful and unjust not to mention the help we have found both in the writings of our respected teacher Mr. Vauquelin and in the advice that his benevolent friendship has continually prompted him to give us over the past twenty years.

The work that we now present to the public consists of six books, which we will first discuss briefly.

#### BOOK I

First of all, this book provides a number of definitions that we think are necessary for a clear understanding of this work. They are drawn from earlier writings and the as yet unpublished monograph mentioned above. Secondly, the book provides a detailed description of the analytical method we used to determine the elemental composition of oils and fats. This method comprises burning the sample with copper oxide, just as Gay-Lussac determined the carbon to nitrogen ratio in uric acid by burning this acid with the same oxide. We would be honored if our analyses of elemental compositions were to be regarded as a continuation of the work of Gay-Lussac and Thenard<sup>10</sup>, and Berzelius. We are bound to acknowledge our great debt to these two French scientists who developed the method of determining the elemental composition of organic compounds.

#### BOOK II

This book describes all the species<sup>11</sup> of fatty matter of animal origin that we have investigated.

These species are described as follows:

1. Composition of the species

After having described the composition of the species that show some acidity in the samples that we studied, we indicate the proportion of water that results from heating that acid with lead oxide. According to the current state of knowledge, one could believe either that this water

How our research on animal oils and fats has been presented in this book

#### **INTRODUCTION**

xxxv

had simply been separated from the acids or that it had been produced by some of their hydrogen and the oxygen in the lead oxide. According to the first hypothesis, the acids as analyzed should be *hydrates*<sup>12</sup>. According to the second one, they are *hydrogen acids*<sup>13</sup>. In this book, we have taken the first hypothesis as our point of departure but with respect to the second hypothesis, we have also given in the Notes the composition of the fraction of the acids that combined with the lead while being heated with the lead oxide. We must admit that the volatilization of the ammonium stearate and palmitate<sup>14</sup> that had been prepared by a dry process whereby no water was liberated<sup>15</sup>, favors the second hypothesis. We also add that lately we have observed quite a number of phenomena in compounds other than edible oils and fats that now look to us to be more in line with the hydrogen acids hypothesis than with the hydrate hypothesis<sup>16</sup>.

2. *Physical properties of the species* 

3. Chemical properties that have bee observed without changing the elemental composition of the species

4. Chemical properties as observed in the species when it is brought into contact with other compounds or conditions that induce a change in its composition

We felt that we should introduce this distinction between two groups of chemical properties rather than describe successively in the same book what is observed when the species are subjected to heat, light, electricity, simple reagents, acids, etc. because there is a link between the properties of a certain group that would be lost if the chemical properties were simply presented one after the other. In fact, it is clear that the properties belonging to the first group are the only ones that can be considered to be an essential part of the nature of the substance because they stem from the kind of nature that remains unchanged by the circumstances under which we observed the phenomena that result from those properties. The properties of the second group also pertain to this nature but they only become evident when it is changed in such a way that it is in fact the elements of the species that react, either on their own or by giving rise to products that are less complex than the species themselves. For example, when water combines with lime<sup>17</sup> and salts, it still acts like water and its nature undergoes no change. But if water vapor is passed over red-hot iron, it decomposes and no longer acts as water but like oxygen<sup>18</sup>.

We regret all the more that we have not had time to study the effect of chlorine and iodine on oils and fats since some phenomena we observed when bringing palmitic acid into contact with chlorine looked extremely interesting<sup>19</sup>. We also regret that we have not determined all the effects that sulfuric acid and especially nitric acid have on oils and fats; we had only expected to find effects that would be characteristic of only a small number of compounds. We would have liked to extend our experiments to confirm our suspicion that an acid that had formerly been

prepared by Vogel by treating 'axonge'<sup>20</sup> with nitric acid is identical to the acid that we obtained from all species of oils and fats in which the atomic ratio of hydrogen to carbon is the same or almost the same as in 'hydrogène percarburé'<sup>21</sup>. If so, this acid would have the same relationship to oils and fats that produce it, as oxalic acid has to substances that release significant quantities of the latter when treated with nitric acid.

- 5. Occurrence of the species
- 6. Preparation of the species
- 7. Nomenclature of the species and synonyms
- 8. History of its discovery and of the work this discovery has led to

#### BOOK III

Now that the main properties of the species of oils and fats have been described, we present appropriate methods to prepare them in the state in which we have studied them. We explain our methods and in a table, we summarize all the steps that may be needed to isolate the saponification products from the most complex fatty matter.

We preferred to devote a separate book to the preparation of most of our species rather than including it in the previous book since our methods are novel and involve a large number of steps

#### BOOK IV

Here we discuss the animal fats that comprise several immediate principles and fats from human cadavers.

#### BOOK V

Having now established the composition of saponifiable oils and fats as well as that of their saponification products, we will now try to verify our analyses by comparing the saponifiable moieties in oils and fats with the saponification products on the basis of their elemental composition. Subsequently, we will consider saponification with respect to alkaline bases in general and more in particular with respect to the proportion of the alkaline base that can saponify a given weight of fat.

#### BOOK VI

This book presents general remarks under different headings. After having discussed the current state of knowledge about oils and fats before we started our work, we consider the immediate principles of

#### INTRODUCTION

which these oils and fats are composed, more or less in the order in which we carried out our investigations. We explain the *principle* to be used to establish what amounts to a compound in organic chemistry. This principle can be seen as the shortest possible summary of our analytical research. It has been our guiding principle for a long time and we hope it will have a most propitious influence on the progress of analytical chemistry by providing young chemists with a *criterion* they have had to manage without until now. Then we will summarize the most general conclusions that can be drawn from our research: 1. with respect to the effect of solvents on the salts of fatty acids; 2. with respect to saponification. We will point out several applications of our work and finally, we will speculate on the immediate principles of which saponifiable oils and fats are composed.

Nearly all the research in this book has been presented to the Academy of Science<sup>22</sup> in the form of reports in chronological order, which is very different from the order in which our findings are presented in this monograph. During the three years that have passed since this book was written, we have done our utmost to make it as worthy as possible of scientists' attention. When more recent work made us suspect that the accuracy of certain experiments could be improved, we did not hesitate to retrace our steps. Accordingly, most of our elementary analyses have been repeated several times. Since we want above all that the truth be known, we openly explain the difficulties we had to overcome and nowhere do we try to hide any gaps that need be filled to make our work complete. Those who would like to continue this work will sense the truth of what we put forward and those who would like to apply it, be it in the arts or in science, will not go far wrong if they carefully read and digest what we have reported.

<sup>&</sup>lt;sup>1</sup> Nowadays, copy-editors would almost certainly delete this negative publicity but fortunately, they did not yet exist in 1823 so that the honesty of the author has been preserved.

<sup>&</sup>lt;sup>2</sup> Here the author refers to himself and his scientific method.

<sup>&</sup>lt;sup>3</sup> These secondary consequences as elaborated by the author are in fact his main contribution to the understanding of the chemistry of oils and fats.

<sup>&</sup>lt;sup>4</sup> In the 18<sup>th</sup> century, chemists supposed that all combustible bodies contained a substance that was released in combustion; they called this substance "phlogiston". Antoine Laurent Lavoisier (1743-1794) demonstrated that combustion leads to a weight gain and prepared oxygen by decomposing mercuric oxide. He thereby refuted the phlogiston theory.

<sup>&</sup>lt;sup>5</sup> Here the author refers to the 'distillative autoclave' he describes in more detail on the next page.

<sup>&</sup>lt;sup>6</sup> This is the first time Chevreul introduces the concept of *principe immediate*; from his definition in Book I Chapter 1, sub-section (5), the term *structural moiety* can be arrived

at as the most likely description. However, the original term 'immediate principle' will be used.

<sup>7</sup> A yellow-flowered plant of the cabbage family, also known as *Isatis tinctoria*, which was widely grown in Britain as a source of blue dye, which was extracted from the leaves after they had been dried, powdered and fermented. (Oxford English Dictionary). See also: M.E. Chevreul, Analyse chimique de l'*Isatis tinctoria* et de l'Indigofère, *Ann. Chim.* **68**, 284-312 (1808).

<sup>8</sup> The author uses the term "éther sulfurique" but according to a book (Charles Moureu, *Notions Fondamentales de Chimie Organique*, Gauthier-Villars, Paris, 1913), which my father used when studying chemistry, this term is synonymous with "Éther ordinaire" or diethyl ether. However, some people held the opinion that the sulphuric acid formed part of the ether and made a distinction between sulfuric ether and nitric ether.

<sup>9</sup> Presumably, some fellow scientists had carried out a few experiments and generated a substance that had a fatty feel. By refuting that any oil was made, the author also highlights the shortcomings of this approach to science.

<sup>10</sup> Chevreul spells this author's name as Thénard, with an acute accent, but there are also authors who omit the accent. When I queried this with Jaime Wisniak, he pointed out that Paul Thenard, son of Louis-Jacques baron Thenard (1777-1857), spells the family name without an accent and he suggested we follow this example.

<sup>11</sup> In endnote 2 on page 4, an explanation is given of the background of the use of the word 'species'. In practice, it often means the same as 'compound'.

<sup>12</sup> The author distinguishes between *hydrates* and hydrogen acids. Sulfuric acid is an example of a hydrate since it can be regarded as hydrated sulfur trioxide just as its salts can be regarded as a combination of a metal oxide and sulfur trioxide. Hydrogen chloride on the other hand, is a hydrogen acid since it contains no oxygen but for quite some time, it was considered to be a compound of oxygen and an undiscovered element, *muriaticum*. Acids had been associated with oxygen since Lavoisier. In German for example, oxygen is called Sauerstoff, which translates as 'acidity matter'.

<sup>13</sup> They could be hydrogen acids but the hydrated acids also contain hydrogen that could react with the oxygen in the lead oxide. In the light of current knowledge, the author's conclusion is not correct, but we should not forget that we now think in terms of hydrogen ions being the cause of acidity. It was Humphrey Davy (1778-1829) who introduced this concept since all acids yielded molecular hydrogen on electrolysis.

<sup>14</sup> Chevreul uses the term "margarate d'ammoniaque", but as explained in the Foreword, the term maragaric acid is now used for heptadecanoic acid (C17:0).

<sup>15</sup> We would now write this reaction as: RCOOH +  $NH_3 \rightarrow NH_4(RCOO)$  and indeed, no water is liberated when ammonium soaps are formed from ammonia.

<sup>16</sup> For a chemist, these were exiting times. Here the author concedes that he changed his mind with respect to acids. Berzelius adopted the new views by 1820.

<sup>17</sup> According to the reaction of calcium oxide (lime) with water:  $CaO + H_2O \rightarrow Ca(OH)_2$  forming slaked lime, water does not stay intact but reacts. Heating this slaked lime causes the reaction to proceed in the opposite direction and thereby liberate water. This may well be the reason why Chevreul assumes the water does not react.

<sup>18</sup> Both water vapor and pure oxygen react with iron under formation of iron oxide, so water and oxygen must have something in common.

<sup>19</sup> Allowing iodine to react with fatty acids would have revealed a difference between saturated and unsaturated fatty acids and in a later stage, between monounsaturated fatty acids having more than one double bond.

<sup>20</sup> In French, the word 'axonge' refers to a fat that is somewhat softer than lard or tallow and a Google Advanced Scholar Search provides the information that lard is used for food and axonge for ointments. It also refers to a patent (this turns out to be U.S. Patent No. 2,51,304) that discloses a process for the randomisation of lard.

<sup>21</sup> A Google Advanced Scholar search for 'hydrogène percarburé' yields a single hit: the current work, which is also the only place where the word 'percarburé' has been used. Chevreul uses it again in sub-section (480) when discussing cetyl alcohol and comparing the water content of cetyl alcohol with that of diethyl ether and ethanol. In that sub-section, he gives the molecular formula of his 'hydrogène percarburé' as (CH<sub>2</sub>)<sub>n</sub>. Accordingly, it will be translated as ethylene.

<sup>22</sup> When he wrote this, Chevreul was not yet a member of the French Academy of Science. He had applied for membership in 1816 but Joseph Louis Proust (1754-1826) was elected instead. When the latter died, Chevreul was elected to succeed him and so became a member on 9 August 1826. He was to preside over the Academy in 1839 and 1867.